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Short Communication

Geogenic lead isotope signatures from meat products in Great Britain: Potential for use in food authentication and supply chain traceability

Jane A. Evans^{a,*}, Vanessa Pashley^a, Gemma J. Richards^b, Nicola Brereton^c, Toby G. Knowles^b^a NIGL, BGS, Keyworth, NG12 5GG, UK^b School of Veterinary Science, University of Bristol, Bristol BS40 5DU, UK^c The Food and Environment Research Agency, Sand Hutton, York YO41 1LZ, UK

HIGHLIGHTS

- Lead (Pb) isotopes measured in modern British meat were geogenic in origin.
- The match indicates that this technique may be used to provenance biological products.
- There was no evidence for a contribution from modern anthropogenic Pb sources.

GRAPHICAL ABSTRACT

Isotope tracing



Lead in meat



Anthropogenic: no



Geogenic: yes

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ABSTRACT

This paper presents lead (Pb) isotope data from samples of farm livestock raised in three areas of Britain that have elevated natural Pb levels: Central Wales, the Mendips and the Derbyshire Peak District. This study highlights three important observations; that the Pb found in modern British meat from these three areas is geogenic and shows no clear evidence of modern tetraethyl anthropogenic Pb contribution; that the generally excellent match between the biological samples and the ore field data, particularly for the Mendip and Welsh data, suggests that this technique might be used to provenance biological products to specific ore sites, under favourable conditions; and that modern systems reflect the same process of biosphere averaging that is analogous to cultural focusing in human archaeological studies that is the process of biological averaging leading to an homogenised isotope signature with increasing Pb concentration.

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* Corresponding author.

1. Introduction

International food scares such as melamine in milk products (China), methanol in spirits (Eastern Europe), horse meat in beef products (Europe), and more recently concerns over Pb levels in noodles (India), illustrate the global nature of the food supply and importance of supply chain traceability and testing as a means of providing assurance in the integrity of our food. Although DNA analysis has a major role to play in distinguishing between species, reliable tests are required that can fingerprint the geographic origins of products entering the human food chain (authentication). In this study, we investigate the potential for the isotopes of lead (Pb), measured in animal products, to act as one such geogenic marker at a local scale.

There are four naturally occurring isotopes of Pb (^{204}Pb , ^{206}Pb , ^{207}Pb , and ^{208}Pb), with the last three being end-members of the natural uranium (U)–thorium (Th) decay series (radiogenic Pb). The usefulness of Pb as a provenance tool lies in subtle, yet measurable, variations in the relative abundances of each of the radiogenic isotopes within ores of different ages. The evolution of these differences is described by the Holmes–Houtermans Model (Faure, 1986). Ores formed at different geological times therefore possess Pb isotope ratios which reflect the growth of radiogenic Pb prior to mineral formation, or ‘closure’. This natural variation in Pb isotopes allows for their use as tracers within environmental systems (Gulson, 2008; Komárek et al., 2008). Pb entering the food chain is not fractionated by biological processes (Dickin, 1995; Faure, 1986; Fowler et al., 1993; Roux et al., 2008), and thus, the ingested/adsorbed Pb ‘signature’ of a sample directly reflects that of its source (Smith and Flegal, 1995). When Pb exposure is derived from interaction with an in situ geological source, this is referred to as geogenic exposure, and the isotope signature of a sample can be used to geographically locate the place of exposure (Montgomery et al., 2010; Reimann et al., 2014) in a manner analogous to the way in which Sr is used as a tracer in archaeological studies (Bataille and Bowen, 2012). Although Sr is ingested through consumption of food, predominately cereals, Pb tends to be taken up through direct ingestion or inhalation of dust and soil particulates (Abrahams and Steigmajer, 2003; Farmer et al., 2011).

Mining has been carried out in Britain for millennia (Johnston, 2008; Juleff and Bray, 2007; McFarlane et al., 2014). Consequently, much of the bioavailable Pb is anthropogenic, which if present in sufficient concentrations, has the potential to swamp naturally available Pb (Kamenov and Gulson, 2014). This was illustrated by Montgomery et al. (2010) who used archaeological samples (teeth) to track the changing Pb isotope signature in humans with time; from Neolithic samples, which reflected a varied geogenic Pb source, through to the Roman and industrial ‘culturally focused’ Pb isotope values, which reflects an homogenised UK signature formed as Pb mined from a variety of UK sources was traded, recycled and reused.

Today, a significant source of anthropogenic Pb in the UK environment remains that which was added to petrol as an anti-knock agent during the 1960s until when it was banned by the EU in 2000 (Bacon et al., 2006; Farmer et al., 2000; Farmer et al., 2010; Farmer et al., 2005; Monna et al., 1997). This ‘petrol Pb’ largely originates from the geologically ancient mines in Australia (Broken Hill and Mt Isa) and hence has a very distinctive $^{206}\text{Pb}/^{204}\text{Pb}$ composition around 16.1 (Gulson, 1986) that is clearly distinguishable from that in geologically younger, Carboniferous aged mine deposits of Britain which has a $^{206}\text{Pb}/^{204}\text{Pb}$ of about 18.4.

Although Pb in petrol was withdrawn in the UK in 2000, a study by Noble et al. (2008) showed that it still makes a significant contribution to the atmospheric Pb budget, with isotope values measured at London air monitoring stations showing only a very slow return to pre-petrol Pb/natural British Pb values.

Our hypothesis was that the Pb isotope signature of the livestock would reflect the animal’s interaction with its environment. Samples in this study were collected from three areas of Pb mineralisation in

Britain; the predominantly Carboniferous mineralisation of the Mendips and Pennines, and the Palaeozoic deposits of, central Wales (Fig. 1). Pb isotope data from these ore fields were available (Barreiro, 1996; Barreiro and Spiro, 1997a; Fletcher et al., 1993; Haggerty et al., 1996b; Moorbath, 1959; Rohl and Needham, 1998) and details of mine locations in Wales are given in Ball and Nutt (1976), providing an excellent geogenic reference for comparison with the samples.

2. Materials and methods

Samples of kidney, liver, or blood from cattle, sheep and hens (broilers and layers) were sampled from the three areas of Britain: Aberystwyth, in Wales, Pennine area of Derbyshire and the Mendip Hills of Somerset in a study commissioned by the UK’s Food Standards Agency (FSA) (Knowles and Richards, 2014). These are historical Pb mining areas and so have the potential to cause elevated levels of Pb in animals. Details of the selection and origins of the samples are given in Knowles *ad bid*.

2.1. Pb isotope analysis

Pb isotope analysis of the samples was conducted using a Nu Instruments Nu Plasma, a Multi Collector-Inductive Coupled-Plasma Mass Spectrometer (MC-ICP-MS). Prior to analysis, each sample was filtered (Millipore 0.25 μm PFA) and spiked with a thallium (Tl) solution, which was added to allow for the correction of instrument induced mass bias. Samples were then introduced into the instrument via an ESI 50 $\mu\text{l}/\text{min}$ PFA micro-concentric nebuliser attached to a desolvating unit (Nu Instruments DSN 100). For each sample, five ratios were simultaneously measured ($^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, $^{208}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{208}\text{Pb}/^{206}\text{Pb}$). Each individual acquisition consisted of 60 sets of ratios, collected at 5-second integrations, following a 60 second de-focused baseline.

The precision and accuracy of the method were assessed through repeated analysis (one standard for every 3–4 unknowns) of an international standard US National Bureau of Standards NBS 981 Pb reference solution (also spiked with Tl). The average values obtained for each of the measured NBS 981 ratios were then compared to the known values for this reference (Thirlwall, 2002). All sample data were subsequently normalised, according to the relative daily deviation of the measured reference value from the accepted reference value. Normalisation to an international standard in this way effectively cancels out the effects of slight daily variations in instrumental accuracy, and allows the direct comparison of the data obtained during different analytical sessions. Internal uncertainties (the reproducibility of the measured ratio) were propagated relative to the external uncertainty (i.e. the excess variance associated with the reproducibility of the reference material analysed during the session). Average 2SD (standard deviation) uncertainties for the ratios are as follows $^{206}\text{Pb}/^{204}\text{Pb} \pm 0.012\%$, $^{207}\text{Pb}/^{204}\text{Pb} \pm 0.012$, $^{208}\text{Pb}/^{204}\text{Pb} \pm 0.015$, $^{207}\text{Pb}/^{206}\text{Pb} \pm 0.004$, and $^{208}\text{Pb}/^{206}\text{Pb} \pm 0.014$. Procedural blanks for chemical separation and mass spectrometry were <100 pg and procedural sample repeats agreed within analytical uncertainty.

Data are presented in the supplementary section and plotted in Figs. 2 and 3.

3. Results

In Fig. 2a ($^{208}\text{Pb}/^{206}\text{Pb}$ vs $^{207}\text{Pb}/^{206}\text{Pb}$) and Fig. 2b ($^{208}\text{Pb}/^{204}\text{Pb}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$) the sample data are plotted together with published Pb isotope data from relevant local ore bodies (Barreiro, 1996; Barreiro and Spiro, 1997b; Fletcher et al., 1993; Haggerty et al., 1996a; Rohl, 1996). The plots highlight different aspects of the data: whereas ($^{208}\text{Pb}/^{204}\text{Pb}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$) plots thorogenic Pb against uranogenic Pb, hence fully separating out the geochemically different decay systems, it is dependent on using the ^{204}Pb ratios (Ellam, 2010). Older



Fig. 1. Sketch map of southern Britain showing the location of the areas of Pb mineralization referred to in this study.

data, generally measured using thermal ionisation mass spectrometry (TIMS), can lead to higher uncertainty on the ^{204}Pb measurement. The ($^{208}\text{Pb}/^{206}\text{Pb}$ vs $^{207}\text{Pb}/^{206}\text{Pb}$) benefits from the higher precision of the larger Pb isotope peaks but may not resolve the data fields as well. We present the data in both forms but focus on the ($^{208}\text{Pb}/^{206}\text{Pb}$ vs $^{207}\text{Pb}/^{206}\text{Pb}$) plot because of our use of older, lower precision, reference datasets.

3.1. Biological samples

Samples from central Wales form two sub-sets of data trending towards higher $^{207}\text{Pb}/^{206}\text{Pb}$ vs $^{208}\text{Pb}/^{206}\text{Pb}$ (Fig. 2a), both of which are clearly distinguishable from the samples of the Mendips and Derbyshire. The data from the Mendip animals forms two clear data groups

in Figs. 2a & b coherent groups whereas the data from Derbyshire is more scattered and occasionally overlaps the upper Mendip group. These results show that the Pb isotope composition of animals from central Wales was clearly distinguishable from the Mendips and Derbyshire reared animals.

3.2. Relationship with Pb isotope data from ore fields

There is a very close match in Pb isotope composition between that measured in the animal samples and published isotope composition of galena (Pb bearing mineral) from the areas in question. The Welsh data show a particularly good match. The ore datasets for central Wales split into two groups, reflecting the slightly higher $^{207}\text{Pb}/^{206}\text{Pb}$ ratios in some of the Welsh locations (Ball and Nutt, 1976; Fletcher

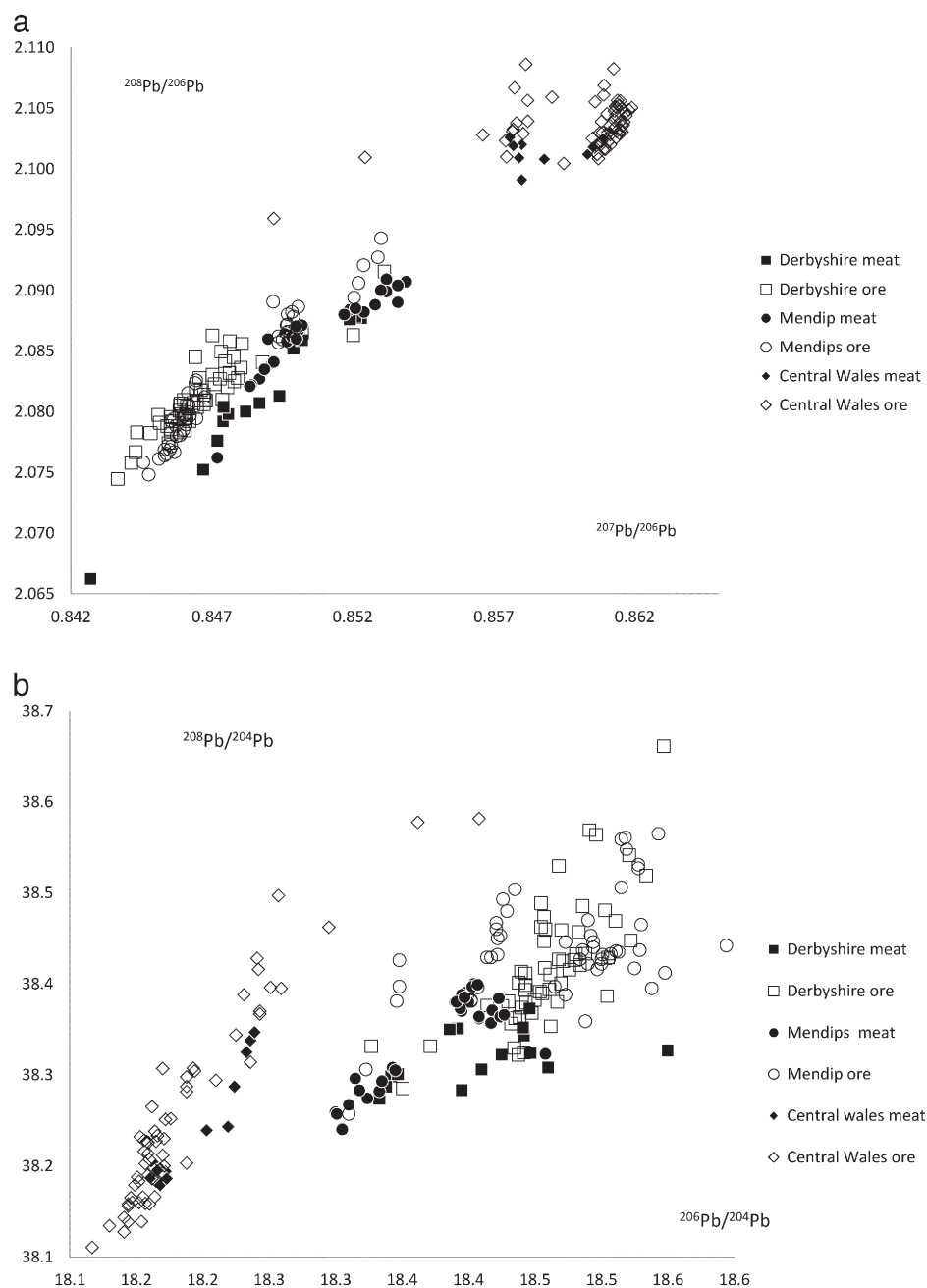


Fig. 2. a & b. Comparison of the Pb isotope composition of meat samples with ore compositions from similar areas of Britain. Ore data from (Barreiro, 1996; Fletcher et al., 1993; Haggerty et al., 1996a; Rohl, 1996). Analytical uncertainty on the ore data approximately $\pm 0.1\%2\text{SD}$.

et al., 1993). The animal data show the same pattern and split into two clear groups that neatly overlap the ore data for the areas.

The biological data from the Mendips also show a strong relationship with the regions ores. The published ore data for the Mendips roughly defines three groups; Mearhead mine ($^{207}\text{Pb}/^{206}\text{Pb}$ c. 0.852); a central group comprising: Charterhouse, Green Ore, Buddles Wood, St Cuthbert's and All Eight's mines ($^{207}\text{Pb}/^{206}\text{Pb}$ c. 0.849), and the remaining locations defining the group with the lowest Pb isotope ratios mines ($^{207}\text{Pb}/^{206}\text{Pb}$: 0.841–0.846) (Haggerty et al., 1996b). One group of the biological samples from the Mendips shows a close association with the Mearhead mine ore values and another group plot within the isotope space as the Charterhouse group of mine ore compositions. No biological samples are associated with the main Mendip ore array, but eight samples plot on the less radiogenic side (higher $^{208}\text{Pb}/^{206}\text{Pb}$ and $^{207}\text{Pb}/^{206}\text{Pb}$) of the Charterhouse group trending towards the low value ore samples.

Data from Derbyshire does not show the close association of values between the biological samples and the ore data. The biological samples plot broadly on a sub parallel trend to the Derbyshire ore data. Four Derbyshire samples compositions plot on the Mendips fields. We can speculate that this offset may be caused by the animals having been purchased from an area of the Mendips and taken to Derbyshire (all samples are thought to be from one farm but confidentiality restrictions prevent confirmation). Equally, however, this overlap may be coincidental as the Mendip and Pennine ore field compositions overlap significantly.

The relationship between the isotope composition ($^{207}\text{Pb}/^{206}\text{Pb}$) and Pb concentration of the biological samples is shown in Fig. 3.

Fig. 3 shows two clear peaks of isotope compositions at approximately $^{207}\text{Pb}/^{206}\text{Pb} = 0.85$ and 0.861. Two reference ranges are displayed on the diagram. The first is taken from 18th–19th century Pb isotope data obtained from human teeth, which represent the

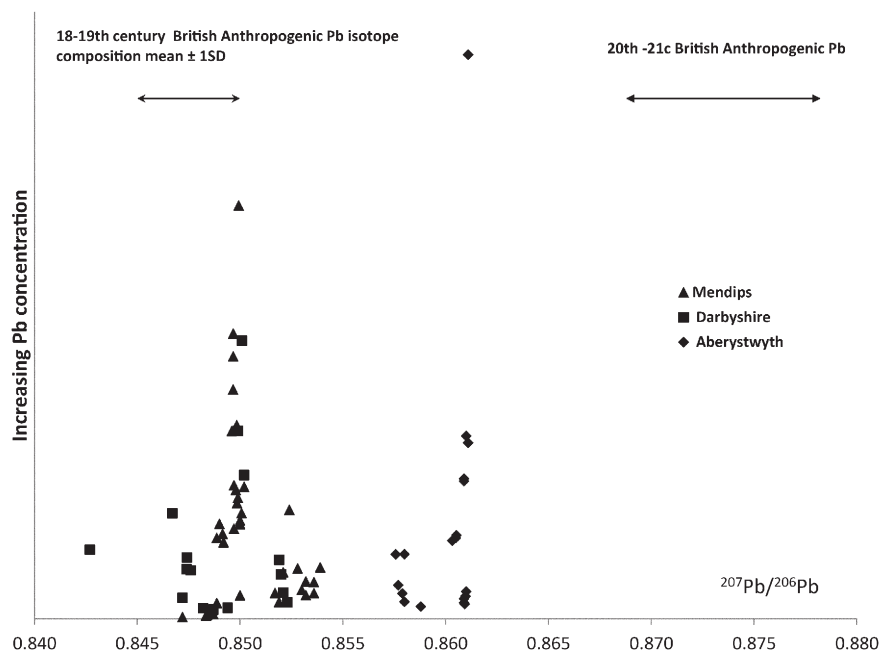


Fig. 3. Cultural focussing diagram showing the tendency to restricted isotope composition with increasing Pb concentration in meat samples. Reference data fields are given as mean and 1 SD (Millard et al., 2014; Noble et al., 2008).

average biosphere uptake of Pb that predates the modern introduction of Australian Pb in petrol (Millard et al., 2014). The second field represents the range of recent Pb isotope measurements of air pollution in London showing the residual effect of the Australian petrol Pb, i.e. modern anthropogenic Pb (Noble et al., 2008). The first peak from the biological samples ($^{207}\text{Pb}/^{206}\text{Pb} = 0.85$) coincides with the typical pre-modern pollution composition seen frequently in British archaeology studies (Montgomery et al., 2010). The second peak ($^{207}\text{Pb}/^{206}\text{Pb} = 0.861$) plots between conventional English values and the field of modern pollution. This second peak is defined by the Welsh data. No data plot within the field of modern anthropogenic values. This diagram shows that although there is a spread in isotope composition within the geographic areas, there is a tendency with increasing Pb concentration towards an average isotope value. Anthropogenic Pb composition of Britain in the 18th and 19th centuries was dominated by Pb derived from Pennine and Mendip ores and hence the co-incidence of many of the samples in this study with this old anthropogenic signature. Modern anthropogenic Pb is dominated by the residual effects of Australian Pb that was used as an anti-knock agent in petrol; its current composition is given by the data of Noble et al. (2008). The tendency of population data to tend towards a singular value with increasing concentration is called cultural focusing when seen in human archaeological datasets (Montgomery et al., 2010) and is caused by the averaging effect of biosphere processes (Reimann, 2008).

4. Discussion

There are a number of studies that demonstrate the imposition of anthropogenic Pb isotope signatures on food products. Studies of wine show variable sources of introduced Pb (Gulson et al., 2006; Medina et al., 2000; Stockley et al., 2003) and the process of making and storing vinegar introduced Pb contamination to a variable extent (Ndung'u et al., 2011). Airborne particulate pollution was shown to influence the Pb isotope composition of wines from the Czech Republic to varying degrees (Mihaljevic et al., 2006). The contribution of anthropogenic Pb from petrol is diminishing in the UK but still exists in measurable form (Noble et al., 2008). However, outside cities and industrial areas, the relationship between environmental Pb concentration and isotope composition with geology shows that “majority of Pb in European

agricultural soils is, at present, largely of natural origin” (Reimann et al., 2012). The data from our study shows that the animals from the Mendips and central Wales have a close and clear relationship to the land on which they were raised/grazed. This not only shows that the process of slaughter and meat processing contributed no measurably different source of Pb, but it also shows that the animals were kept in a clean environment away from sources of modern anthropogenic Pb.

5. Conclusion

This study highlights three important observations; that the Pb found in modern British meat from these three areas is geogenic and shows no clear evidence of anthropogenic Pb; that the generally excellent match between the biological samples and the ore field data, particularly for the Mendip and Welsh data, suggests that this technique might be used to provenance biological products to specific ore sites, under favourable conditions; and that modern systems reflect the same process of biosphere averaging that is analogous to cultural focusing in human archaeological studies.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.scitotenv.2015.07.133>.

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